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The twice-renormalized Rouse formalism of polymer dynamics: Segment diffusion, terminal relaxation, and nuclear spin-lattice relaxation

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Abstract

The twice-renormalized Rouse formalism, a refined version of Schweizer's renormalized Rouse treatment of chain dynamics in entangled polymers, is presented. The time scale of validity is extended to include the terminal chain relaxation and center-of-mass diffusion. In clear contrast to the laws concluded from other polymer dynamics concepts (such as the reptation (tube) model or the polymer mode-mode coupling formalism), the predictions perfectly coincide with all the results of recent spin-lattice relaxation dispersion and diffusion experiments as well as with computer simulations. On the other hand, the twice-renormalized Rouse formalism fails to explain the rubber-elastic plateau of stress relaxation. It is inferred that this is a consequence of the single-chain nature of the present approach not accounting for the fact that viscoelasticity is largely a manifestation of collective multichain modes. In the rigorous sense, no such multichain treatment has yet been established to our knowledge. The necessity to consider interchain cooperativity in any real comprehensive polymer dynamics theory is concluded from low-frequency spin-lattice relaxation data, which are shown to reflect fluctuations of long-distance intermolecular dipole-dipole interactions.